The investigation of the forced piston effect at various densities of pure fluids around the critical point $^{\! 1}$

C. Bartscher², J. Straub^{2,3}

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² Institute A for Thermodynamics, Technical University Munich, Boltzmannstrasse 15, 85748 Munich, Germany.

³ To whom correspondence should be addressed.

ABSTRACT

In recent years the research of critical phenomena started to focus on the dynamic non equilibrium behavior of critical fluids which includes the investigation of the "piston effect" or "critical speeding up". For this purpose a new sample cell with volume changing capabilities was designed to investigate a density spectrum of $\pm 50\%$ of the critical density ρ_c of pure sulfur hexafluoride (SF₆) and measure the influence of the average fluid density on the critical fluid behavior.

Up to now the piston effect is studied only where a pressure wave type expansion change of a cooled or heated boundary layer creates a volume change (the "piston"). With the volume changing mechanism, realized by means of a real movable piston, a "forced piston effect" can be introduced in a defined and measurable way into the fluid during the process of adjusting the fluid density in the sample cell. Three thermistors observe first a fast then a subsequent temperature relaxation. Additionally a pressure transducer indicates the pressure behavior.

The cell is integrated in a Van der Waals-Zeeman thermostat known from various Space Shuttle and MIR missions. The temperature control and data acquisition is handled by the sophisticated French ALICE 2 facility which is made available for microgravity and ground based experiments, due to a German-French co-operation contract. The scientific objectives of this research project and first preliminary results from ground based reference experiments are presented.

KEY WORDS: critical phenomena, density variation, forced piston effect, sulfur hexafluoride

1 INTRODUCTION

In the past fifteen years several studies of critical phenomena have been performed on ground and under microgravity using sounding rockets, facilities on spacelab during shuttle missions and on the MIR station. In most of these experiments the sample cell was filled with SF_6 of constant critical or near critical density. SF_6 was used as test fluid because of its convenient and proper critical parameters.

We have now designed a sample cell in which the density of the fluid can be varied in a range of $\pm 50\%$ of the critical density ρ_c . With this we can study the influence of various average densities on the behavior of a fluid near its critical state, both on ground and under microgravity. The volume changing mechanism has been realized by means of a movable piston. As the mass of SF_6 in the cell is constant, the density variation is inversely proportional to the volume change.

Up to now the piston effect studied theoretically and experimentally by serveral authors [1,7,8,9,10] could be seen in a finite volume when the boundary layer was exposed to a sudden temperature change. This causes either a fast expansion or contraction of the boundary layer creating a shock wave propagating through the fluid resulting in a quick temperature dissemination.

In our cell the expansion and contraction of the boundary layer, corresponding to a volume change, can be simulated with the piston movement performed in a defined measurable way. In this case we talk about the "forced piston effect".

2 EXPERIMENTAL SETUP

The experimental setup consists of a sample cell unit incorporated in a thermostat which can either be inserted in the French ALICE 2 facility or a laboratory setup with temperature control and data acquisition.

2.1 THE INSTRUMENT

The ALICE 2 (Analyse des LIquides Critiques dans l'Espace) is an advanced French facility [5] for the investigation of phase transitions and mass and heat transport phenomena of fluids near their critical point under microgravity conditions. The facility is built around a thermostat unit that provides up to two fluid samples with a highly stable thermal environment. Fluid behavior is monitored through accurate temperature and pressure measurements at low and high sampling rates, and through optical diagnostics, such as microscopy, direct observation, and grid shadow technique. A central data management system performs automatically all the commands and controls of a preprogrammed experiment cycle. Temperature and pressure data as well as facility house-keeping data are stored and CCD images are recorded. A separate electronics is specifically dedicated to the thermal control of the thermostat containing fluid samples.

2.2 THE THERMOSTAT

For obtaining a stable temperature environment a well-tried thermostat from the Van der Waals-Zeeman Laboratory of the University of Amsterdam, is used. Similar types flew successfully during various missions on the space shuttle (IML-1 and 2 in CPF) and on MIR in the facilities ALICE 1 and 2. Figure 1 shows the slightly modified

three shell thermostat containing a system of heater foils, peltier elements and temperature probes to guarantee a very stable temperature environment. The thermostat can either be inserted in the Alice 2 facility, which contains the temperature control, all optical diagnostics, and the data storage system or used in the laboratory with a Van der Waals temperature control and a self-made data acquisition system. The temperature in the sample cell unit (SCU) can be controlled in both cases to a stability better than ± 0.1 mK. To keep losses from heat conduction and the temperature gradient in the SCU as low as possible most parts of the valve and piston mechanism are made of material with a small heat conduction coefficient.

The SCU, made of aluminum (thermal diffusivity $a=93.27*10^{-6}\frac{m^2}{s}$), incorporates the $sample\ cell\ (\approx 1.4\ cm^3)$ filled with SF_6 and a variable $compensation\ volume$, both connected with each other over a thin canal. The canal can be closed with a $valve\ mechanism$ to separate $sample\ cell$ and $compensation\ volume$ from each other. The size of the $compensation\ volume$ can be changed by means of a piston manipulated mechanically with a handwheel. It allows a density variation of the test fluid in a range of $\pm 50\%$ of the critical density ρ_c . A turn indicator helps to define the magnitude of the volume/density change up to a accuracy of $\Delta\rho$ =0.4% ρ_c . When the valve mechanism is turned, however, the volume change is always fixed at ΔV =4.922 mm³, which makes also small but defined density changes possible.

2.3 THE SAMPLE CELL

Figure 2a shows a sketch of the sample cell and the positions of the measurement thermistors in the cell. The cell has a shape of a cylinder with 12 mm in diameter and

length respectively. *Thermistor 1* (Th-1) is positioned 1 mm off the center of the cell, thermistor 2 (Th-2) is located very close to the wall and 6 mm below Th-1, and thermistor 3 (Th-3) is mounted also very close to the wall, but outside the cell so that it has no direct contact with the fluid. In the laboratory the cell can be rotated by 180°, so that depending on the filling one of the inner cell thermistors is always in the liquid and the other one in the gaseous phase. A piezoresistive pressure sensor is used to detect pressure changes in the fluid. In this configuration the only optical analysis tools are direct observation and grid shadow technique.

The *canal* between the two volumes can be closed with a valve after the fluid's density has been changed in order to avoid any disturbing influences from the compensation volume on the test fluid which could not be observed, particularly during phase transition and relaxation experiments. For the forced piston effect experiments, however, the valve is left open and therefore ignored in this figure for the reason of simplicity.

3 SCIENTIFIC OBJECTIVES

The basic idea of the piston effect is that a sudden rise in temperature in the wall of a sample cell with a constant volume causes a thermal expansion of the fluid in the boundary layer. This results in a compression of the bulk fluid and an average pressure increase [4]. Assuming negligible temperature gradients outside the boundary layer the compression is adiabatic and under the assumption that no energy is dissipating the compression can be regarded as isentropic, too, leading to a temperature increase. Therefore the energy transport to the center of the cell is a mechanical process, propagating like a

pressure wave, defined by the speed of sound w_s and the isentropic compressibility χ_s [2].

In our experiment we try a different approach. According to Figure 2a one wall of the compensation volume consists of a movable piston which translatorical movement is very well defined, as is the subsequent volume change. Under the assumption of a homogeneous temperature distribution as an initial condition, all three thermistors in the sample cell see the same temperature $(T_1=T_2=T_3)$, identical to the wall temperature T_W . T_W is above the coexistence curve in the one-phase region of the fluid and kept constant with the thermostat temperature control.

Moving the piston by a well defined length the volume is changed by ΔV and therefore the average density by $\Delta \rho$ (Fig. 2b). This creates a pressure wave propagating through the cell inducing a real "piston effect", what we call the "forced piston effect". If the average density change $\Delta \rho$ is performed fast enough and under the assumption of negligible temperature gradients and no energy dissipating, the compression is adiabatic and the temperature change is equal in the bulk fluid and is nearly isentropic.

Due to the capability of measuring temperature and pressure changes (ΔT , Δp) at a high resolution the difference coefficients can be regarded as differential coefficients when the step size of $\Delta \rho$ is adapted accordingly and the piston movements are performed rapidly at a well defined length. Hence, with $\left(\frac{\partial p}{\partial \rho}\right)_s$, $\left(\frac{\partial T}{\partial \rho}\right)_s$ or $\left(\frac{\partial p}{\partial T}\right)_s$ several isentropic coef-

ficients can be calculated.

From the temperature and average density change the isentropic expansion coefficient α_s can be determined with

$$\alpha_s = -\frac{1}{\rho} \left(\frac{\partial \rho}{\partial T} \right)_s, \quad \left(\frac{\partial \rho}{\partial T} \right)_s \sim \frac{\Delta \rho}{\Delta T}$$

as well as the isentropic compressibility χ_s from the subsequent average change in pressure Δp with

$$\chi_s = \frac{1}{\rho} \left(\frac{\partial \rho}{\partial p} \right)_s, \quad \left(\frac{\partial \rho}{\partial p} \right)_s \sim \frac{\Delta \rho}{\Delta p},$$

and with Δp and ΔT the isentropic tension coefficient β_s

$$\beta_{s} = \frac{1}{p} \left(\frac{\partial p}{\partial T} \right)_{s}.$$

With these isentropic coefficients further thermodynamic relations can be derived and used as a check of the consistence of thermodynamic equations, such as the speed of sound

$$w_S = \rho \chi_S^{-\frac{1}{2}}$$

or the isochoric specific heat capacity

$$c_V = -\frac{\rho}{pT} \alpha_S \beta_V$$
 with $\beta_V = \frac{1}{p} \left(\frac{\partial p}{\partial T} \right)_V$.

The isochoric tension coefficient β_v can be measured from isochoric changes of T with the same apparatus. While the temperature is crossing the coexistence curve, the dynamic process of the phase separation can also be observed as long as the experiments are performed not too close around the critical point. Within the region of about (T-T_c)= ± 0.5 K and $\Delta \rho = \pm 15\%$ of ρ_c , however, the effect of gravity on the fluid's behavior is

dominating too much and, therefore, the near critical region should be investigated in space under microgravity conditions.

These density steps can be performed in the same way in the 2-phase region. In this case, measuring the temperature rise simultaneously in both phases the isentropic tension coefficient β_s can be obtained. While β_s in the gaseous phase is larger than in the liquid phase it should converge to the same value when the critical temperature T_c is reached.

After a density variation the temperature in the bulk fluid has changed to a different value, while the temperature of the wall, controlled by the thermostat, is kept constant on the initial value. Therefore the fluid will return to the initial temperature with an exponential function, according to theory [6]. From this the relaxation time τ of the system can be derived, which is defined as

$$\tau = \frac{R^2}{D_{th} * C}$$

where R is a characteristic length of the cell, D_{th} the thermal diffusivity of the fluid, and C a geometrical constant. If R and C are determined in a fluid region, where D_{th} is known, the relaxation process can be used to determine D_{th} close to the critical point as a function of T and ρ . In the two-phase region with the knowledge of the thermal diffusivity of both phases at saturation, the influence of the interface on the mass and heat transport across the interface can be estimated.

4 PRELIMINARY RESULTS

Figures 3a, 3b, and 4 depict some effects caused by density changes performed during first preliminary ground-based experiments to test the facility not for scientific evaluation. No matter whether the density is increased or decreased Th-1 (see fig. 2) is always exposed to a greater change in temperature than Th-2 which is influenced by the constant wall temperature $T_w = T_3$.

Figure 3a shows the effect of a 0.22% volume increase (density decrease !) performed with the *valve mechanism* (see chapter 2.2). The experiment starts at an average density ρ =1.34 ρ_c , with a constant wall temperature T_w = T_c +2K and homogeneous temperature distribution (T_1 = T_2 = T_3 = T_w). Almost immediately with the start of the volume change the pressure wave reaches the thermistors Th-1 and Th-2 and both observe a steep temperature decrease. T_1 drops to almost -65 mK and T_2 to -40 mK, both relative to the wall temperature T_w , which remains constant within the measurement resolution of Δt_{min} =0.6 mK.

The discontinuity of the temperature decrease is the result of a two-step volume change indicating that the fluid is responding very fast to the density variation. Immediately after the volume remains constant both curves return to the initial temperature $T_{\rm w}$ exponentially. This is supported by the fact that after being cooled by the forced piston effect the fluid touches the warmer wall and experiences an expansion due to the temperature gradient and the thermal piston effect starts. Remarkably, the curve of thermistor 2 is flattening after 2 seconds and even crossing the thermistor 3 curve after 13 seconds. This has to be investigated in more detail, but one explanation can probably be found in the gravity effect in combination with the position of the thermistors in the cell

(fig. 2). Th-1 is located 6 mm above Th-2 and experiences a larger temperature drop leading to a higher local density in the fluid. As a result, conviction starts and the colder fluid sinks to the bottom of the cell (Th-2). After long enough period of time (not shown in the graph) all temperatures converge to the initial, which is kept constant by the thermostat temperature control.

The volume change in figure 3a starts with the same temperature conditions, whereas the average density is at $\rho=\rho_c$. This time the volume is <u>decreased</u> by 0.17% in 3 steps. As expected, both inner cell thermistors observe immediately a steep temperature increase, thermistor 1 reaching a peak of $\Delta T=30$ mK and thermistor 2 a $\Delta T=25$ mK. It is obvious that the absolute temperature change is smaller due to the smaller volume change. Again, as soon as the volume change is stopped, both temperatures start to decay immediately. While T3 is returning to the initial constant temperature T_w exponentially, the relaxation process of T2 reveals quite interesting dynamics. First the gradient is much steeper, second a quite noticeable undershoot below the initial temperature can be observed.

The same effect is seen in figure 4. Here the density is increased quite drastically in 13 steps from ρ =1.15 ρ_c to 1.4 ρ_c at a set point temperature of T_{set} = T_c +200 mK. Due to the huge density variation the temperature in the cell is rising by more than 500 mK, which makes it impossible to keep the wall temperature T_w constant. T_w even increases by more than 50 mK before it returns to the initial value due to the temperature control (not shown here). Therefore in this graph, the temperature changes of T_2 and T_3 are relative to the set point temperature used as the reference temperature.

It can clearly be seen that T2 undershoots not only the wall temperature but also the initial set point temperature. This observation coincides with the observation of [3] who observed in his experiments that local fluid temperatures can undershoot their initial value after a heat pulse occurs. This is an indication that the piston effect behaves the same or at least similar, no matter whether it was generated thermally by a heat pulse from a thermistor or mechanically with a sudden volume change.

This temperature undershoot will be studied further together with the observation of the density relaxation by means of interferometry, which will also give information about the convection process due to induced local temperature gradient generated from a mechanical piston effect.

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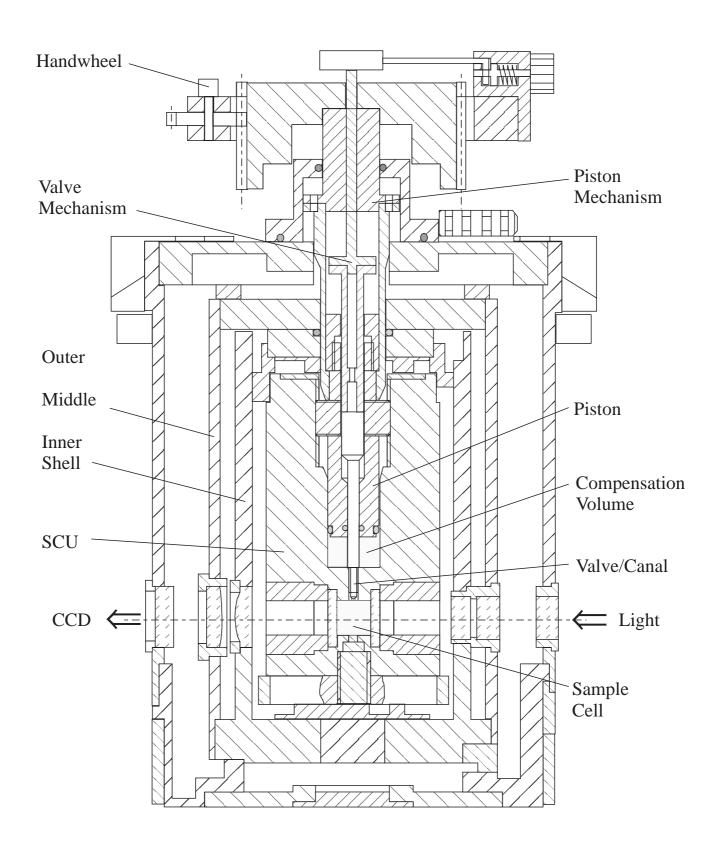
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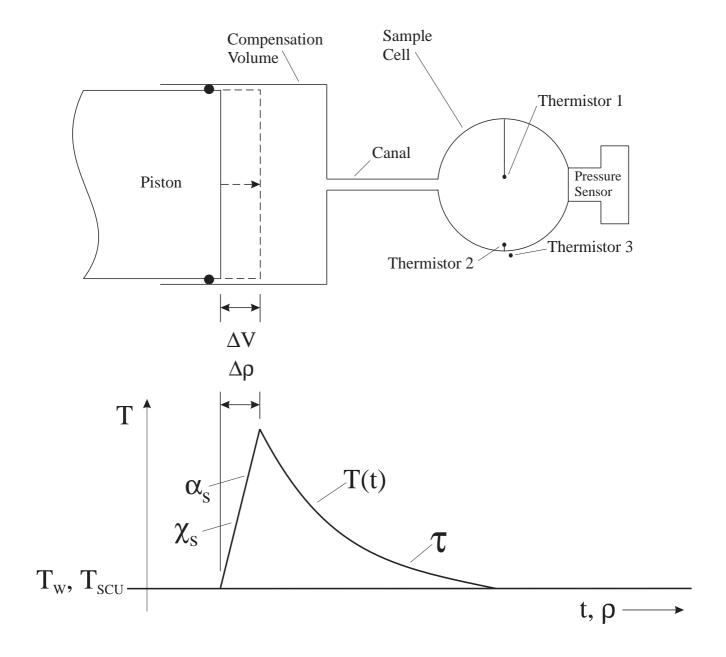
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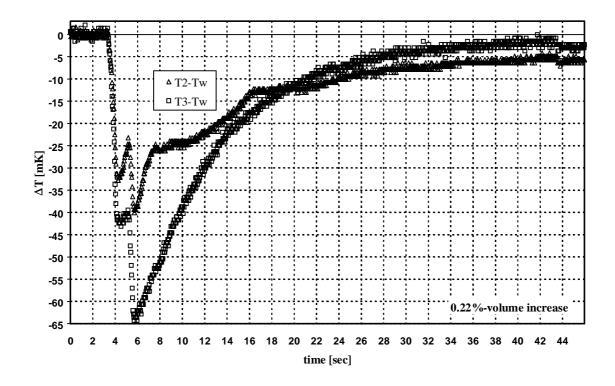
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FIGURE CAPTIONS

- Fig. 1. Sketch of thermostat with integrated sample cell unit (SCU) and volume changing mechanism
- Fig. 2. Detailed cell illustration with measurement sensors and principle of volume change. Temperature behavior after density variation.
- Fig. 3. a) Temperature changes relative to constant wall temperature T_w = T_c +2 K at TH-1 and TH-2 after 0.22% volume decrease. Initial density is ρ =1.34 ρ_c . b) Temperature changes relative to constant wall temperature T_w = T_c +2 K at TH-1 and TH-2 after 0.17% volume increase. Initial density is ρ = ρ_c .
- Fig. 4. Temperature changes relative to set point temperature T_{set} = T_c +200 mK during a density increase from 115% to 140% ρ_c in 13 steps.







b)

